

## INTRODUCTION

Youngstown State University's Isomer Physics Project is a program of basic physics research with the goal of providing credible, high-quality measurements and theoretical analysis related to achieving a controlled release of energy stored in nuclear isomers. An emphasis has been on providing a firm foundation coupling traditional nuclear structure research, such as from spectroscopic studies, with focused triggering experiments. As such, success is defined not by proving or disproving specific preconceptions, but by obtaining the best possible insight into physical reality. This reality can then be used to consider possible applications for particular isomers. Under the present contract, and as part of the first year of the DARPA SIER Program, effort was concentrated on examining the possibility of triggered gamma emission from the 31-year isomer of  $^{178}\text{Hf}$ . This effort was divided into three distinct areas, two experimental and one theoretical.

Possibilities for triggered of gamma emission from  $^{178\text{m}2}\text{Hf}$ , with concomitant energy release, may be naturally divided into two energy domains. The first is so-called "low-energy" triggering with incident photon energies less than 100 keV. This is the sole energy domain which has been investigated to-date, although there is strong concern as to the validity of those experimental results that provide evidence in support of triggering. The YSU Isomer Physics Project aimed to conduct high-quality measurements of low-energy triggering.

The second energy domain is that of so-called mid-energy triggering, with incident photons of 100 keV – 2 MeV. Although less likely to be of value for triggering in an application due to the higher incident energy, this range might be important for depletion of isomers by the incidental photon flux during production cycles. Established nuclear spectroscopy of the  $^{178}\text{Hf}$  nucleus shows a 126-keV transition which will support triggering – a trigger experiment has not yet been conducted. Also, recent publications [1, 2] indicate that other trigger transitions at 330, 990 and 1,676 keV may exist – these have not yet been tested in a triggering experiment, but initial system development was conducted under this contract. A third energy domain, so-called "high-energy" triggering for incident photon energies greater than 2 MeV is not a subject of present study.

In addition to the two experimental regimes, theoretical analysis was conducted to evaluate the likelihood that the established NEET process [3] could provide a mechanism for low-energy triggering as suggested in reports of positive evidence of triggering (as summarized in [4]).

## SCIENTIFIC BACKGROUND

Nuclear isomers have served as subjects of study for decades, since their first observation [5] in 1921. Appearing throughout the periodic table, the existence of "long-lived" excited nuclear states reflects specific aspects of nuclear structure [6] that provide a barrier to the decay of those levels. This may come about due to a true potential barrier such as between different nuclear shapes, leading to shape isomers like the  $T_{1/2} \sim 14$  ms

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14. ABSTRACT Youngstown State University's Isomer Physics Project is a program of basic physics research with the goal of providing credible, high-quality measurements and theoretical analysis related to achieving a controlled release of energy stored in nuclear isomers. An emphasis has been on providing a firm foundation coupling traditional nuclear structure research, such as in spectroscopic studies, with focused triggering experiments. As such, success is defined not by proving or disproving specific preconceptions, but by obtaining the best possible insight into physical reality. Under the present contract, and as part of the DARPA SIER program during its first year, effort was concentrated on examining the possibility of triggered gamma emission from the 31-year isomer of <sup>178</sup> Hf. This effort was divided into three distinct areas, two experimental and one theoretical. This report describes results obtained in these areas. At this point, no support was found for claims of triggering of <sup>178m2</sup> Hf near 10 keV.					
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level of  $^{242}\text{Am}$ . More typically isomers exist due to angular momentum considerations as for the archetype spin isomer (spin trap), the  $J^\pi = 9^-$  level lying at 75 keV in  $^{180}\text{Ta}$ . Since an M8 transition would be required for electromagnetic decay to the ground state (with similar inhibition for electron capture and beta decay), this spin isomer possesses a halflife in excess of  $10^{15}$  years [7]. It is noteworthy that the conclusion that the long-lived level was an excited one rather than the ground state came only in 1980 [8]. The difficulty in understanding the stellar nucleosynthesis of this rarest isotope of the rarest element has motivated considerable study (see [9] and references therein). The production mechanism was a major issue, but also important was the question of whether there was any way an isomeric population could be depleted through inelastic photon scattering within a star [10].

The depletion of the  $^{180}\text{Ta}$  isomer was first observed in 1988 [11] although the specific transitions from the metastable level to intermediate states identified later [12-14]. The accepted model for this process involves two steps: first, an incident photon is absorbed by the  $^{180}\text{Ta}$  nucleus in the isomeric state, being excited into a so-called intermediate state. Second, the intermediate state decays with some branch leading by cascade to the ground state. Experimentally, the convenient lifetime of the ground state (8.2 hours) allowed straightforward detection of depletion in out-of-beam gamma-ray measurements. No ground state exists naturally, so observation of any decay of that level after irradiation of a sample signifies depopulation of the isomer. The lowest-lying intermediate state is about 1 MeV above the isomer [13] and a tentative correlation has been made between it and a level found from spectroscopic studies [15]. At present, it appears that photon-induced depopulation of  $^{180\text{m}}\text{Ta}$  would allow survival of this isotope under some stellar conditions [9] although the question remains open for further study. It should be noted that initial concerns that the strengths of some depletion reactions, through specific intermediate states, were contrary to established nuclear systematics [16] have been ameliorated by later results [17].

While studies continue on  $^{180\text{m}}\text{Ta}$ , the only "free" isomer,\* interest in depletion reactions has expanded to other isotopes [18]. Principle among these is the K isomer,  $^{178\text{m}2}\text{Hf}$ . For deformed nuclei such as  $^{178}\text{Hf}$ , the additional, approximate, quantum number K is introduced as the projection of the total nuclear angular momentum on the body axis, thus quantifying the relative orientation of that vector. Decay rates for even dipole transitions may be significantly reduced if there is a need for large K (orientation) changes between levels – this would be exemplified by transitions having multipolarity  $L < \Delta K$ . These are not truly forbidden, as would be the case for  $L < \Delta J$ , but are instead strongly inhibited [19] and this can lead to modest lifetimes. If an intrinsic state of high angular momentum is yrast, then no decay transitions of dipole character will be available and higher multiplicities are required. Coupled with any K hindrance, this can lead to quite long-lived states. These effects account for the 31-year halflife of the 2.446-MeV,  $J^\pi = 16^+$  state in  $^{178}\text{Hf}$ . Its electromagnetic decay proceeds primarily through an E3 transition at 12.7 keV. Between the initial and final states, however,  $\Delta K = 8$  so that the required excess orientation change (degree of K forbiddenness)  $\nu = \Delta K - L = 5$ . This

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\* From stellar nucleosynthesis; excludes isomers occurring in natural Th and U chains.

strongly reduces the decay rate from that obtained from a Weisskopf estimate to the observed value by an empirical factor of about  $(100)^5$  [19], giving an extremely long halflife – strong internal conversion ( $\alpha = 1.39 \times 10^7$ ) gives the measured 31-year halflife of this K isomer,  $^{178m2}\text{Hf}^\dagger$ . An essential point for this and other isomers near  $A \sim 180$  is the existence of high- $\Omega$  single-particle orbitals near the Fermi surface, leading to high-K intrinsic states. In this particular case, the isomer is a four-quasiparticle state [20] with configuration  $\nu^2 7/1 [514]9/2^+ [624] \otimes \pi^2 7/2^+ [404]9/2^- [514]$ . This intrinsic level is yrast, being more than 990 keV below the  $16^+$  member of the ground state band [21].

It was suggested [22, 23], based on systematics studies of  $(\eta, \gamma')$  on isomeric nuclei, that it might be possible to depopulate  $^{178m2}\text{Hf}$  in a similar way to that found in  $^{180m}\text{Ta}$ . The first published report [24] of a so-called “triggering” experiment gave statistically weak evidence in support of the detection of isomer depletion using a modest bremsstrahlung source. The lower bound [24] on the integral cross section for the process was on the order of  $10^{-21} \text{ cm}^2 \text{ keV} = 10^6 \text{ eV b}$ , very much larger than would be expected from established nuclear systematics. This issue was raised in several comments [25-27], but clearly the lesson of  $^{180m}\text{Ta}$  indicates that further experiments might well resolve this contradiction. The canonical trigger energy reported from that initial experiment [24] was 40 keV. A number of subsequent experiments have been performed and published, with continued positive reports from the group of Collins, et al and null measurements being reported by all other groups. A summary of the literature up to late 2003 may be found in Ref. [4].

The results of Collins, et al (see publications listed in [4]), if correct, challenge our understand of not only nuclear physics, but also the interaction of nuclei and atomic electrons. Due to the extremely large cross sections deduced from their data, it was suggested that pure nuclear photoabsorption did not initiate triggering. Instead, the process of nuclear excitation by electron transitions (NEET) was invoked. Here, the first stage involves the creation of a vacancy in an inner electron shell and, of course, the cross section for this is many orders-of-magnitude larger than that of nuclear photoabsorption at the same energy. Then, provided sufficient matching exists between a nuclear transition and an electron transition (to fill the vacancy), some atoms would undergo NEET as energy was transferred by a virtual photon into the nucleus rather than in emission of X-ray fluorescence. The process is generally well-established [3], although the probability of NEET is typically quite small – the only case in which NEET has been conclusively observed is that of  $^{197}\text{Au}$  [28]. In that work, the so-called NEET probability (ratio of measured cross section for the NEET process to the photoelectric cross section for the relevant atomic shell) was found to be  $5.0 \times 10^{-8}$ , in good agreement with modern calculations.

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<sup>†</sup> The 31-year state is the second long-lived metastable state, thus m2. A lower lying metastable,  $^{178m1}\text{Hf}$ , has a halflife of 4 s.

## LOW-ENERGY TRIGGERING STUDIES FOR $^{178\text{m}2}\text{Hf}$

Several experimental series were performed at the BL12B2 Taiwan contract beamline at SPring-8 during June 2003. The research team consisted of personnel from YSU, the Joint Institute for Nuclear Research (Dubna, Russia), SRS Technologies, Inc., and Taiwan's National Synchrotron Radiation Research Center (NSRRC). The experiments by the YSU/Dubna/SRS/NSRRC team at SPring-8 were designed to test the claims of Ref. [29] with improved statistical accuracy and greater signal-to-noise ratio.

It has been reported ([29], SIER Kick-off and May 2003 HIPP meeting presentations by Collins) that the most compelling evidence (largest effect) for triggering was from an enhancement to the numbers of 213-keV gamma rays from  $^{178\text{m}2}\text{Hf}$  which occurred when an isomeric target was irradiated with monochromatic radiation at SPring-8 near 9.568 keV. In Ref. [29] a Ge detector of unspecified efficiency was used to obtain gamma spectra from the target during irradiation periods of 50 s (continuous acquisition). The counts in the 213-keV line for incident energies below the  $L_3$  edge (9.558 keV) were used to establish a baseline or null-effect condition. Small increases in the areas of the 213-keV line during certain irradiations at energies above the  $L_3$  edge, with the largest enhancement near 9.568 keV, were used to infer the occurrence of triggered gamma emission.

In the June 2003 YSU/Dubna/SRS/NSRRC experiments, the isomeric sample consisted of material originating from LANL [30] and containing 0.36  $\mu\text{Ci}$  activity ( $1.9 \times 10^{13}$  atoms) of  $^{178\text{m}2}\text{Hf}$  and 0.56  $\mu\text{Ci}$  activity ( $1.2 \times 10^{12}$  atoms) of  $^{172}\text{Hf}$  at the time of the experiment, as determined by gamma counting at SRS Technologies, Inc. The hafnium was deposited on a 1-mm thick Be disk having a diameter of 1.905 cm. Characterization of the material properties may be found in Ref. [31]. The hafnium deposit was estimated at time of deposition to have a nominal diameter of 5 mm. The deposit was estimated at the time of deposition to be roughly uniform by a visual inspection, not suffering from the annular features of some other samples. The actual distribution of hafnium atoms was determined as described below. A second 1-mm thick Be disk was placed over the hafnium deposit and an Al assembly was used to press together the disks to provide appropriate encapsulation of the hafnium material into the beam target.

Target was attached to the end of an aluminum arm which in turn was attached to a translation stage. The stage was moved in a step-wise fashion such that the target was raster scanned in a vertical plane across a beam of monochromatized SR at the BL12B2 beamline. The beam of SR was produced by a standard Spring-8 bending magnet and monochromatized by a double Si monochromator. The crystals were rotated in order to defocus the beam to achieve a full vertical dimension of 1 mm and a corresponding spread of bandwidth to 2 eV (employed during the entire experiment), although collimating jaws were used for this raster scan to restrict the beam spot to  $0.5 \text{ mm} \times 0.5 \text{ mm}$ . This size allowed sufficient spatial resolution of the 2-d scan without sacrificing too much experimental time. For this scan, the monochromator was set to transmit photons at a nominal energy of 9,561 eV ( $\pm 0.5$  eV) which corresponds to the  $L_3$

edge of hafnium. SPring-8 standard ionization chambers were positioned to sample the beam before and after passing through the target. Total charge produced within the pre- and post-target ionization chambers during specific dwell periods, reflecting the beam fluence, could be compared to determine which features in scans were due to beam fluctuations and which were due to attenuation in the target. Figure 1 shows the results of the 2-d scan with 0.5 mm resolution obtained with the above-described beam. The spatial scan was automated and charge was collected from the ionization chambers for a dwell time of 1 s at each position. The post-target ionization chamber current has been plotted in a fashion so that the grey scale reflects the correct interpretation of transmission of the target as viewed looking toward the beam. The Al assembly was sufficiently thick to essentially stop the beam at this energy and appears dark (no transmission). The Be transmits most of the beam and is white, and the hafnium deposit with its modest attenuation appears clearly as located within the window of transmission through the Al assembly. It should be noted that the target was inclined, thus the roughly

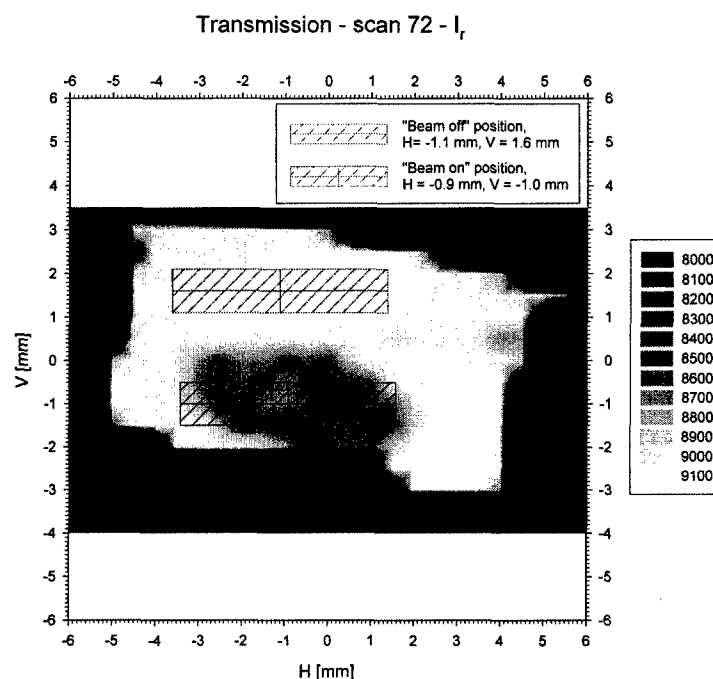


Figure 1: Spatial scan of the inclined target using a 0.5 mm  $\times$  0.5 mm beam spot of monochromatic radiation, showing the amount of transmission through different parts of the sample from the post-target ionization chamber. The target was translated so that gamma spectra could be obtained with the same incident radiation for "on deposit" and "off deposit" conditions.

circular hafnium deposit appears elongated. An important aspect of this figure is that the target rather than the beam is scanned. The roll of the deposit from the horizontal is due to the orientation of the aluminum arm that connected the target to the translation stage. Due to the positioning of various gamma-ray detectors and the attachment of target to the

arm, the arm was not perpendicular to the beam axis and so inclination of the target to the beam introduced this small roll.

Having identified the hafnium deposit within the target and calibrated the positioning of the translation stage relative to the beam, the target was moved so that the  $0.5 \text{ mm} \times 0.5 \text{ mm}$  beam was fixed at specific positions Fig. 1. Holding the target stationary in the beam, the monochromator was rotated in a step-wise fashion to scan in energy across the hafnium  $L_3$  edge. The scan was automated, dwelling for 1 s at each energy and advancing in steps of 1.5 eV. The post-target chamber, notwithstanding any variations in the beam also apparent in the pre-target chamber, reflects the transmission of hafnium with the dip occurring at 9,561 eV. This corresponds well with the known  $L_3$  absorption edge of hafnium. The shape of the transmission dip of Fig. 2 agrees with the expectation of a strong "white line" in the near-edge profile seen in other elements near  $Z = 70$ , although the line cannot be resolved from the edge nor other fine structure seen due to the bandwidth of the beam. Energy scans were performed at a variety of locations, including missing the hafnium deposit but within the Be "window." Scans were also performed at various locations across an energy range covering the  $L_4$  edge of hafnium with expected results. It was determined from the depth of the transmission dips at different locations that the dark (lowered transmission) areas in Fig. 1 did indeed correspond to areas of highest hafnium concentration. Also, little hafnium was to be found outside the elliptical boundary of the deposit.

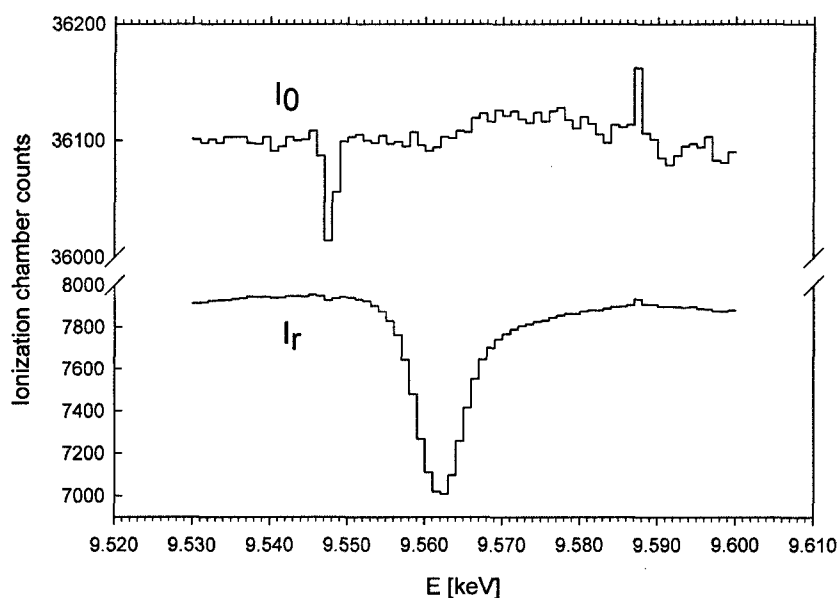


Figure 2: Energy scan over the  $L_3$  edge of hafnium, with a  $0.5 \text{ mm} \times 0.5 \text{ mm}$  beam spot positioned to irradiate the concentration of material at coordinates of (0.28 mm, -1.03 mm)

It should be noted that there was no means available by which to determine if the distribution of hafnium atoms, determined by beam attenuation, accurately reflected the distribution of  $^{178\text{m}2}\text{Hf}$ . There is, however, no physical reason for believing that these distributions are not identical or effectively so.

It is trivial to show that the highest signal-to-background ratio in triggering experiments with  $^{178\text{m}2}\text{Hf}$  is obtained by having the highest possible fraction of isomeric atoms within the beam spot. For this purpose, the target was inclined for this reason and the beam collimators were opened to provide a beam with vertical and horizontal dimensions of 1 mm and 5 mm, respectively. In this way, it was possible to arrange an "on deposit" position of the target which provided an irradiation of hafnium atoms as shown in Fig. 1. An "off deposit" position was determined for the target which irradiated a spot as also shown in Fig. 1. It was determined that no detectable amount of hafnium was irradiated with the target was in the "off deposit" position while 42% of the total hafnium within the deposit was irradiated when the target was in the "on deposit" position. Further inclination of the target was found to require reduction in the beam's horizontal dimension – the chosen combination of inclination, beam dimensions and irradiation positions was optimal for the available target.

Two types of experimental series were employed. The first type was based on a desire to repeat with greater statistical precision the measurements of Ref. [29] in which positive evidence of triggering was reported. Specifically, it was reported that an increase in the numbers of 213-keV gamma rays, corresponding to the  $4^+ \rightarrow 2^+$  E2 transition near the bottom of the gsb, occurred when incident monochromatic SR was scanned across a range of energies from 9,530 eV to 9,630 eV. The increase was maximum just above the  $L_3$  edge as seen in Fig. 5 of Ref. [29]. A similar enhancement of gamma emission due to purported triggering was claimed near the  $L_1$  edge and it was stated that the effect near this edge was "The most pronounced evidence for NEET..." Still, later reports concentrated on evidence of enhanced gamma emission near the  $L_3$  edge and the maximum of the effect was stated to be at 9,567 keV ([32] and numerous SIER presentations). The first experimental series performed here was an energy scan across a range of 9,550 – 9,580 eV as a test of the claim of enhanced emission near the  $L_3$  edge at 9,561 eV.

Gamma spectra were obtained by a 65% P-type Ge detector and were acquired for dwell times of nominally 10 minutes at each fixed monochromatic energy of the incident SR beam and the range of 9,550 – 9,580 eV was scanned in steps of 1.5 eV. This acquisition time was  $\sim 10\times$  that employed in Ref. [29], but our detector was located at a distance of 8.5 cm from the target<sup>†</sup> compared with 2.5 cm in that experiment [29], which

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<sup>†</sup> The 65% Ge detector used to obtain the data discussed here could not be placed closer than 8.5 cm since the target was held by the positioning arm within the center of the YSU miniball detector array. The design of this array is described elsewhere [33, 34] and allows the acquisition of  $\gamma$ - $\gamma$  coincidence (so-called "doubles") and higher-order data, leading to effective calorimetry of gamma bursts from radioactive materials. A transportable and modest-cost alternative to such major arrays as gammasphere, it was emplaced at the beamline as a test of operation prior to additional, future (non-SIER) experiments. The 65% Ge used to obtain "singles" spectra in this work viewed the target through a gap between miniball detectors.



used one Ge detector of 10 mm thickness  $\times$  16 mm diameter<sup>§</sup>. The work of Ref. [32] employed two Ge detectors of 43 – 47 mm thickness  $\times$  45 – 50 mm diameter, each placed 2.2 cm from the sample.\*\* Overall, the statistical accuracy of the present experiment (with 10-minute dwells) may be expected to be at least 4.5 $\times$  that of Ref. [29] and  $\times$  that of Ref. [32].

The work of Refs. [29, 32] compared the emission of gamma rays from a sample during and without irradiation and spectra obtained in the latter condition were taken to establish a “baseline” condition as a zero-effect background. In the present experiment it was possible to provide two forms of backgrounds both of which should reflect a zero-effect condition. First, gamma spectra could be obtained from the target without its irradiation (not allowing SR into the radiation “hutch”). Second, gamma spectra could be obtained from the target while it was positioned “off deposit.” The latter provides a better comparison in that any incident radiation scattered toward the detector from the Be substrate would be identical whether hafnium was being irradiated or not. (Note that the “on deposit” and “off deposit” positions were carefully examined to determine that no Al was struck, which would have caused increase scattering of the incident beam).

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<sup>§</sup> Reference [29] gives no information about detector efficiency, but the provided Ge crystal dimensions correspond to an Ortec GLP-16195/10 P-type planar germanium detector [35]. Efficiencies of such a small detector cannot be given in the standard fashion, relative to a 3 in  $\times$  3 in NaI(Tl) crystal for the 1,332-keV line of <sup>60</sup>Co – the efficiency of the planar Ge detector will be quite small above 400 keV but this does not necessarily reflect negligible efficiency below that energy. Calibration of a significantly larger Ortec GLP-36360/13 at YSU using point sources at 10 cm gives 0.077 % absolute full-energy-peak efficiency at 213 keV. One may expect that the smaller detector used in Ref. [29] at the closer distance of 2.5 cm would provide an efficiency at 213 keV that is comparable or less than that from the YSU GLP-36360/13. In turn, this is less than the 1.3 % absolute full-energy-peak efficiency at 213 keV provided by the “65 %” Ge detector used in this work at 8.5 cm from the target.

\*\* Reference [32] gives no information about detector efficiency, but the provided Ge crystal dimensions correspond to Ortec GMX-10190 N-type coaxial germanium detector, consistent with detectors described in Ref. [36].

Prior to beginning the above-described experimental series, a long background spectrum was obtained of gamma rays emitted from the target in order to establish the performance of the acquisition system and to insure the identification of all gamma rays observed in the "hutch." Figure 3 shows this spectrum obtained over 24,043 s (clock time) with a dead time of 2.2 %. All 171 peaks within the spectrum were positively identified with very few corresponding to radionuclides other than  $^{178\text{m}2}\text{Hf}$  and the daughters of  $^{172}\text{Hf}$  decay. For gamma rays emitted by the hafnium decays, peaks were

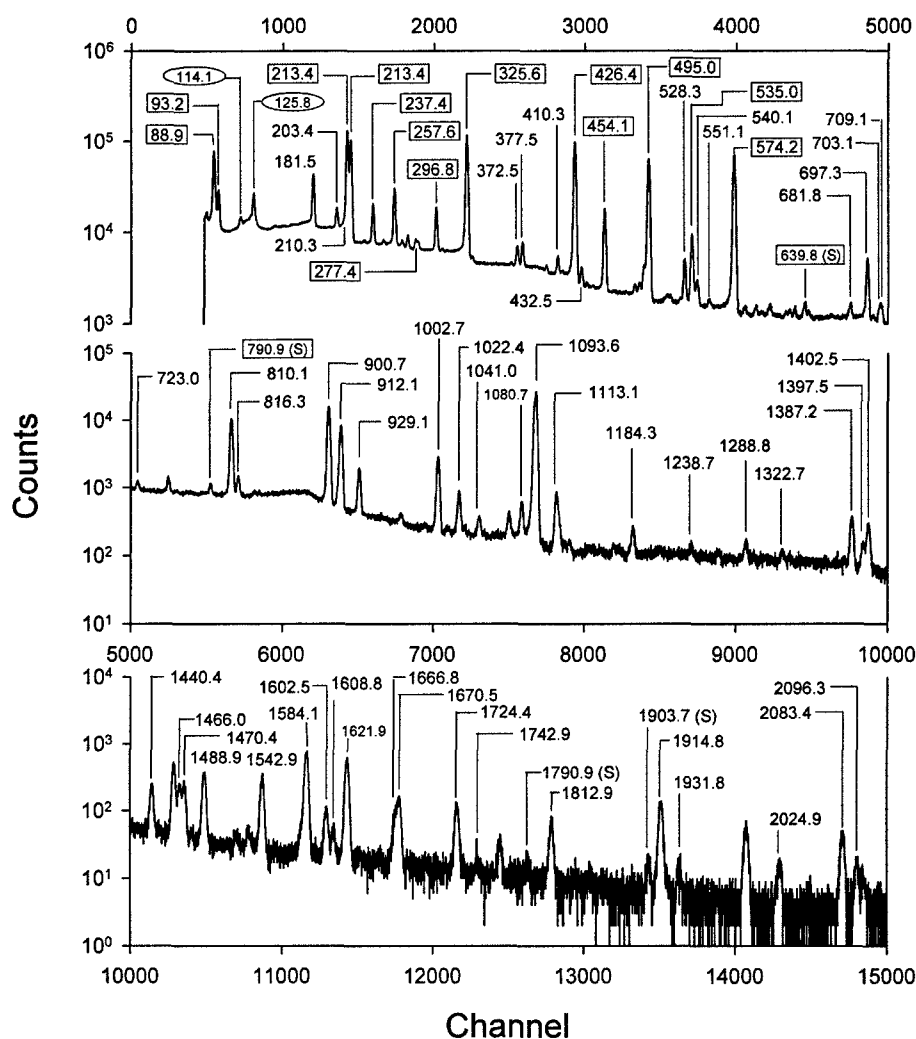


Figure 3: Long background (no beam) gamma spectrum used to identify all contributions due to radioactive hafnium species and natural background in the experimental hutch. All peaks were positively identified, although not all are marked here. Energies (in keV) enclosed in rectangles indicate  $^{178\text{m}2}\text{Hf}$  gammas, ovals indicate  $^{172}\text{Lu}$  gammas and bare entries are for  $^{172}\text{Yb}$  gammas.

positively identified not only by energy but also by relative intensity after the peak areas were scaled by the detector efficiency. It is worthwhile to note that for gamma rays from  $^{178\text{m}2}\text{Hf}$  spontaneous decay, all transitions found in the ENSDF [37] were identified with the exception of the heavily-converted 12.7-keV  $16^+ \rightarrow 13^-$  transition. Newly-discovered, weak transitions [38] in the  $^{178\text{m}2}\text{Hf}$  natural decay were not observed. Gamma peaks resulting from transitions within decay daughters from  $^{172}\text{Hf}$  were observed down to an intensity of 0.037 % per  $^{172}\text{Hf}$  decay for  $^{172}\text{Yb}$  transitions and down to 0.113 % per  $^{172}\text{Hf}$  decay for  $^{172}\text{Lu}$  transitions. Peak areas were obtained by the commercial FitzPeaks program [39] which was found to be preferable to other programs due to its straightforward ability to provide initially automated peak search and fitting followed by manual inspection of fits when dealing with many spectra. Fits obtained with FitzPeaks were compared with those obtained with the well-known TV [40] and GammaVision [41] packages. Systematics differences were seen between the peak areas from the various programs, but these were quite small and appeared in a consistent manner: for example, peak areas in the spectrum of Fig. 3 determined with FitzPeaks were always slightly smaller than those obtained with TV and so use of the FitzPeaks program would not introduce any inconsistent systematic error in the consideration of differences between peaks obtained during vs. without irradiation. The absolute detector efficiency was measured at YSU's XEL<sup>2</sup> laboratory later using calibrated point sources, but under identical geometrical conditions to those employed at BL12B2. The absolute efficiency at 213 keV was 1.3 % and the efficiency curve was verified by the intensity matching between lines of Fig. 3.

Scanning across the  $L_3$  edge as described above, spectra were obtained with beam on with the target in both "on deposit" and "off deposit" positions, and with no beam entering the hutch with the target in both positions. Figure 4 shows raw areas (counts) for the 213-keV gamma ray peaks in those spectra which were all taken for the same clock durations. Dead times were about 2.5 %, similar to that found in the long background obtained with no irradiation – the live times of the spectra from which the data of Fig. 4 were obtained differed by no more than 0.01 %. To guide the eye, lines show three standard deviations of the mean. No significant variations in the numbers of 213-keV gamma rays are visually apparent between any of the spectra.

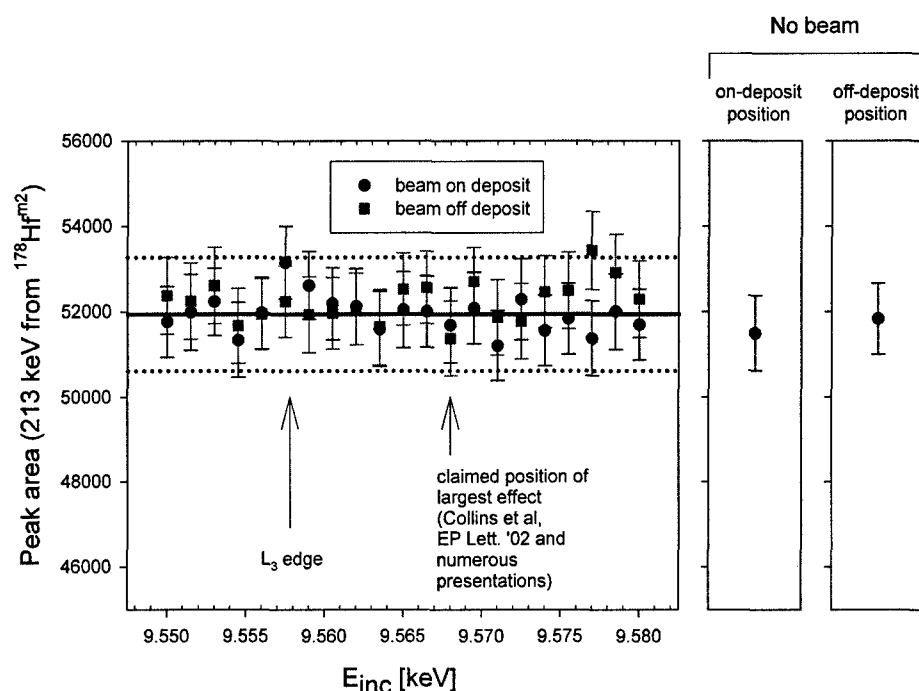


Figure 4: Comparison between 213-keV peak counts in “on deposit” and “off deposit” positions for energy scans across the L<sub>3</sub> edge (and location of largest reported effect). Also included are “no beam” counts for the target in both positions. The solid red line gives the mean value of the measurements, with dotted red lines giving  $\pm 3\sigma$  limits of the mean.

Statistical analysis of the data shown in Fig. 4 was performed using the commercial SPSS [42] to quantitatively examine any variations. The simplest measure is the mean and it was found that a statistically-significant difference existed between the mean of the 213-keV peak counts obtained with irradiation and that corresponding to the counts without irradiation. The mean of 213-keV peak areas across the energy scan were 0.5% larger from the un-irradiated spectra. This small difference may be simply attributed to the small change in solid angle from hafnium deposit to detector when the target was translated between the two irradiation positions. Means were also obtained in a similar fashion from the raw peak areas for other gamma rays from  $^{178\text{m}^2}\text{Hf}$  decay. The general trend was supported, with some gamma rays providing means with no statistically-significant difference and a few (like 213 keV) providing marginally-significant differences. The average difference between means from “on deposit” and “off deposit” spectra was small and, again, it must be stressed that beam “off deposit” spectra were slightly larger due to the small difference in solid angle.

Further analysis was performed on the scan data from the 213-keV transition. Each set of data (“on deposit” and “off deposit”) was separately analyzed by the unpaired t test to determine if the data were part of a normal (thus random) distribution. Peaks

areas over the scanned energies from the 213-keV gamma ray in the "on deposit" position were found to be part of the same normal distribution at the 95% confidence level. Peak areas for this gamma ray for the "off deposit" spectra were found to be part of the same normal distribution at the 95% confidence level. The two distributions were not the same, differing in the mean as described above. A statistically-significant variation in the peak areas, perhaps due to enhanced emission at this gamma energy near an incident photon energy of 9,567 eV would cause the entire set to not correspond to random deviations from the same normal distribution.

Also, the "on deposit" and "off deposit" sets were separately examined using the unpaired t test as applied to progressive 4-point subsets of the data as compared with the mean of the full set. For example, the mean of the first 4 points as a subset of the "on deposit" set of 213-keV peak areas was compared with the mean of the full set of "on deposit" data. It was determined at a 95% confidence level that these means indicated data coming from the same normal (random) distribution. The subset of points taken were then advanced to the 2<sup>d</sup> through 5th points and the analysis repeated until all possible contiguous 4-point subsets were analyzed. No statistically significant variation of subsets was seen (from the same normal distribution at the 95% confidence level). A statistically-significant variation for some subsets would have been expected if an enhanced emission as reported in Refs. [29, 32, 43], spanning a range of about 100 eV, existed.

A similar energy scan over the range of 11,257 – 11,287 eV, thus covering the L<sub>1</sub> edge at 11,271 eV, also provided no evidence in support of enhanced gamma emission due to triggering.

In all cases, peak search and fitting was first performed automatically and then followed by manual inspection of all spectra. In particular, the spectra were inspected for the appearance of additional peaks, such as near 130 keV as first reported in Ref. [36] and subsequently in Ref. [43]. No such peak was evidenced in any spectra from the current experiment. As a final check of any variations between spectra, the total counts were obtained over the full energy range of every gamma-ray spectrum (about 75 – 2,250 keV). Variations in the total spectral counts were at the level of 0.18 % and at the 95% confidence level indicated that the variations were purely random.

To provide a focused search for evidence of enhancements to gamma emission under irradiation, a second series of experiments was performed. This also employed the 65% Ge detector in a similar fashion to that described above. Spectra were acquired for 30 minutes, dwelling for that duration at only two energies for the incident monochromatic radiation: 9,000 eV and 9,567 eV. The purpose was to obtain a comparison between gamma-ray emission under irradiation at the energy purported to cause the greatest enhancement (9,567 eV) and at an energy well below any L edge of hafnium. The energy of 9,000 eV was chosen for the latter measurement. For each choice of incident energy, spectra were obtained with the target in the “on deposit” and “off deposit” positions. Figure 5 shows the resulting raw peak areas for all relevant gamma peaks from  $^{178\text{m}2}\text{Hf}$  decay. The counts are not statistically different at the 95% confidence level.

Comparison between beam on- and off-deposit spectra  
 $^{178\text{m}2}\text{Hf}$  gamma lines

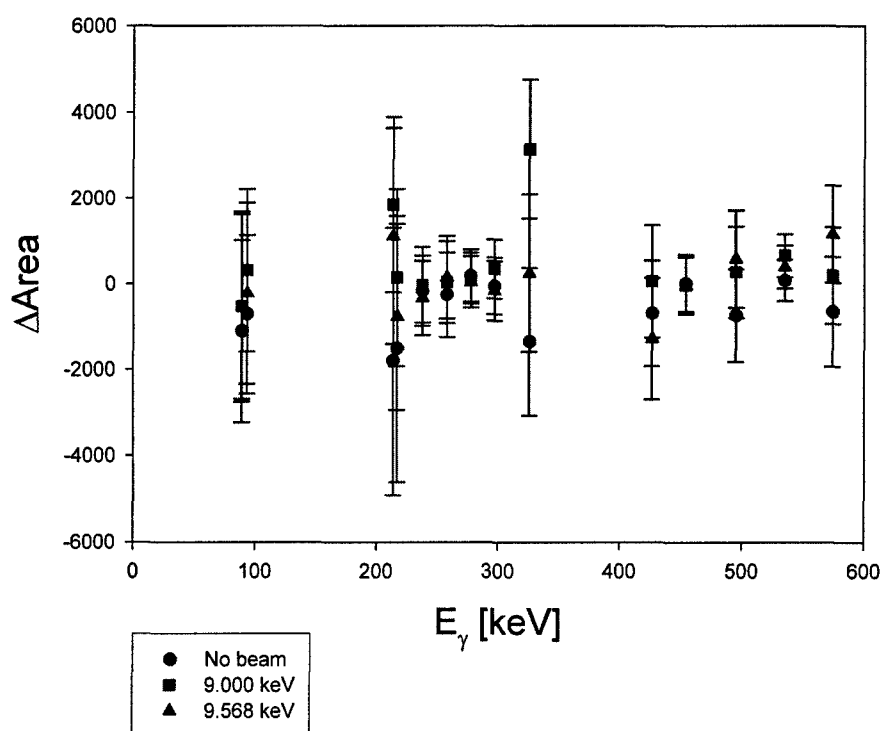


Figure 5: Comparison of peak counts from  $^{178\text{m}2}\text{Hf}$  gamma lines obtained with “on deposit” and “off deposit” irradiations with 9.000-keV and 9.568-keV monochromatic synchrotron radiation, and with “no beam” conditions at both positions.

A follow-up experiment, performed under a separate AFOSR grant (non-DARPA SIER) was conducted during June-July 2004 at the BL12B2 beamline utilizing the YSU miniball detector array to search for evidence in support of low-energy triggering of a prompt nature. Only natural decays from  $^{178\text{m}2}\text{Hf}$  and  $^{172}\text{Hf}$  daughters were observed.

### MID-ENERGY TRIGGERING

Initial tests were performed to begin design of experiments. Due to the lack of monochromatic photon sources of sufficient intensity in this energy range, bremsstrahlung would be required for irradiations. Such experiments are inherently difficult in that the vast majority of the broad-band photon spectra are not utilized for nuclear or atomic excitation and therefore create a large background of ambient scattered radiation. Initial tests indicate that future experiments under other contracts will be near the limit of detectability.

### NEET PROBABILITIES FOR LOW-ENERGY TRIGGERING OF $^{178\text{m}2}\text{Hf}$

Reference [29], and subsequent presentations and preprints, proposed that the process of nuclear excitation by electron transitions, NEET, provided the mechanism for low-energy triggering. Clearly a mechanism that was more exotic than pure nuclear absorption of incident photons was needed since the reported cross section was far too large to be understood on the basis of established nuclear physics. The YSU SIER Project has also aimed to perform a theoretical analysis of the NEET probability for low-energy transitions in  $^{178}\text{Hf}$  and its possible impact on triggering. The analysis, performed by post-doc Michael Harston, one of the world's leading experts in NEET and related electron-nuclear interactions, shows that the probability of NEET in the  $^{178}\text{Hf}$  system is strongly dependent on the B value of the nuclear transition, not as has been supposed by some on a constant "4-Rydberg" value. The upper limit on the NEET probability for nuclear transitions from the isomer to higher-lying levels up to 80 keV above the m2 state was found to be less than  $10^{-8}$  per atomic photoionization, compared with the reported value [29] of  $2 \times 10^{-3}$ . This result, computed by the same techniques which provide excellent agreement with the experimental measurements for NEET in  $^{197}\text{Au}$  [28], was recently confirmed by independent calculations [44]. This latter work assumed perfect matching between electronic and nuclear transitions, and thus is less exact than work performed under the YSU project.

A preprint (from the proof copy) is attached as Appendix A of the associated manuscript in press for Laser Physics 2005. This preprint discusses the details of the calculation and the results, which, in short, indicate that NEET is rather poorly applicable to the  $^{178}\text{Hf}$  nucleus.

### PUBLICATIONS FOR WORK CONDUCTED UNDER GRANT

"Nuclear Excitation and De-excitation in Resonant Electronic Transitions," M. R. Harston and J. J. Carroll, Laser Phys. 14, 1452 (2004).

"Limits on Nuclear Excitation and Deexcitation of  $^{178}\text{Hf}^{m2}$  by Electron-Nucleus Coupling," M. R. Harston and J. J. Carroll, *Laser Phys.* 15, 487 (2005).

"Search for low-energy triggered gamma emission from  $^{178m2}\text{Hf}$  at SPring-8," J. J. Carroll, S. A. Karamian, N. Caldwell, T. Drummond, J. Lazich, J. Lepak, J.-J. Lee, K. Liang, R. Propri, H. Roberts, M.-T. Tang, P. Ugorowski and A. Warner, in preparation for *Phys. Rev. C* (2005).

### EDUCATIONAL IMPACT

Although beyond the typical level of research conducted at comprehensive undergraduate institutions like Youngstown State University, this project has benefited tremendously from the involvement of a number of students at the sophomore, junior and senior levels, and from several different departments. Students Nathan Caldwell, Tom Drummond, Joe Lazich, Joel Lepak, Ron Propri have contributed to every aspect of the planning, design and construction of detector systems and benefited greatly from the unique experience on this project. Their experience is certainly unique at this stage of their careers.

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## APPENDIX A

# Limits on Nuclear Excitation and Deexcitation of $^{178m2}\text{Hf}$ by Electron–Nucleus Coupling

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**Abstract**—Nuclear excitation in transitions that are resonant with electronic deexcitations are discussed in the context of recent claims of the triggering of the decay of  $^{178m2}\text{Hf}$  by irradiation with low-energy photons (C. Collins *et al.*, Laser Phys. Lett. **14**, 154 (2004)). The upper limits for the NEET cross sections are  $\sigma < 10^{-31} \text{ cm}^2$  for transitions filling L-shell holes. These results are significantly below the values of  $10^{-22} \text{ cm}^2$  reported by Collins *et al.* and confirm the recent results by Tkalya (E. Tkalya, Phys. Rev. C. **68**, 064611 (2003)). These calculations suggest that NEET is unlikely to be the mechanism responsible for the reported apparent enhancements of  $^{178}\text{Hf}$  nuclear line intensities by X-ray irradiation.

## 1. INTRODUCTION

There has been considerable interest in recent years in the investigation of low-energy triggering of the decay of the 2.45-MeV isomeric state  $^{178m2}\text{Hf}$  [1]. Null results have been obtained by several groups [1]. Collins and coworkers [2, 3] have, however, reported enhancements in the decay rate corresponding to a cross section for triggering by photons with energies close to the L-shell edge, that is,  $2 \times 10^{-3}$ , the photoionization cross section. The suggested mechanism for triggering was the NEET process, i.e., the excitation of the nucleus by resonant deexcitation of an initially excited electronic state. This process has been detected in the excitation of  $^{197}\text{Au}$  following irradiation by photons with energies just above the K-shell edge. The probability for NEET excitation after creation of a K-shell hole was found to be  $5.0 \pm 10^{-8}$  [4]. Initial theoretical estimates [5] indicated a significantly larger theoretical NEET probability, but a more recent calculation [6] gives a NEET probability of  $3.6 \times 10^{-8}$ , in reasonable agreement with experiment.

The principal object of this article is to obtain estimates of NEET probabilities and cross sections for various possible NEET transitions in  $^{178m2}\text{Hf}$  in order to examine the claims of low-energy triggering in this nuclide.

## 2. THEORY

Let us consider the deexcitation of an initially excited electronic state  $i$  in an atom with the nucleus in level  $I$ . In most cases, this state deexcites to a lower lying state by a radiative or Auger transition. In the case of a NEET transition, the electronic state deexcites by the exchange of a virtual photon with the nucleus, which is excited to the level  $E$ . Symmetry requires that the electronic transition have the same multipolarity as

that of the nuclear transition. The probability for the decay of the electronic state by NEET as opposed to purely electronic decay is known as the NEET probability. This probability is given by

$$P_{\text{NEET}}(i \rightarrow f) = \left(1 + \frac{\Gamma_f}{\Gamma_i}\right) \frac{V_{if}(\pi L)^2}{(\omega_e - \omega_N)^2 + (\Gamma_i + \Gamma_f)^2/4}, \quad (1)$$

where  $\Gamma_i$  and  $\Gamma_f$  are the total widths of the initial and final ( $f$ ) hole states;  $\omega_e$  and  $\omega_N$  are the energies of the electronic and nuclear transitions, respectively; and  $V_{if}(\pi L)$  is the NEET matrix element. Here and below, we use relativistic units ( $c = m = \hbar = 1$ ) unless otherwise stated. The matrix element is given by

$$V_{if}(\pi L)^2 = 4\pi\alpha\omega_N^{2L+2} \frac{(C_{j_i,1/2L0}^{j_f/2})^2}{L^2[(2L+1)!!]^2} |M_e(\pi L)|^2 B_{I \rightarrow E}(\pi L), \quad (2)$$

where  $\alpha$  is the fine structure constant,  $j_i$  and  $j_f$  are the total angular momenta of the initial and final electronic subshells,  $B_{I \rightarrow E}(\pi L)$  is the reduced nuclear transition probability for a transition from the ground nuclear state to the excited nuclear state,  $M_e(\pi L)$  is the electronic matrix element for the transition, and  $C_{j_i,1/2L0}^{j_f/2}$  is a Clebsch–Gordan coefficient. The nuclear electromagnetic moment is here related to the reduced nuclear matrix element by

$$B_{I \rightarrow E}(\pi L) = \frac{|\langle j_E \| M(\pi L) \| j_I \rangle|^2}{(2j_I + 1)}, \quad (3)$$

**Table 1.** Level properties for low-lying electronic levels of hafnium

Subshell	Energy, keV	Width, eV	Photoexcitation cross section, $\times 10^{-20} \text{ cm}^2$
1s	65.351	42	0.28
2s	11.271	5.6	1.1
2p <sub>1/2</sub>	10.739	5.0	2.1
2p <sub>3/2</sub>	8.561	4.9	4.6
3s	2.600	12.7	3.0
3p <sub>1/2</sub>	2.365	4.5	5.1
3p <sub>3/2</sub>	2.108	6.5	14.8
3d <sub>3/2</sub>	1.716	1.5	12.5
3d <sub>5/2</sub>	1.662	3.0	8.5

where  $M(\pi L)$  is the electromagnetic transition operator [7] and  $j_i$  and  $j_E$  are the spins of the ground and excited nuclear states, respectively.

The cross section for production of a given initial atomic state followed by a NEET process can be calculated by summing over NEET transitions that fill the given hole state:

$$\sigma_{\text{NEET}}(i) = \sigma_{PE}(i) \sum_f P_{\text{NEET}}(i \rightarrow f), \quad (4)$$

where  $\sigma_{PE}(i)$  is the cross section for photoexcitation of the hafnium atom, yielding the initial electronic hole state  $i$ .

Once an excited level  $E$  is populated by NEET, it may either decay back to the initial nuclear state  $I$  or to another nuclear level. The experimentally observable

cross section for the population of a nuclear level  $F$  by excitation from the level  $I$ ,  $\sigma_{I \rightarrow E \rightarrow F}$ , is given by

$$\sigma_{I \rightarrow E \rightarrow F}(i) = \sigma_{\text{NEET}}(i) R_{I \rightarrow E \rightarrow F}, \quad (5)$$

where  $R_{I \rightarrow E \rightarrow F}$  is the branching ratio for decay to the state  $F$ :

$$R_{I \rightarrow E \rightarrow F} = \frac{\lambda_{E \rightarrow F}}{\lambda_{E \rightarrow F} + \lambda_{E \rightarrow I}}, \quad (6)$$

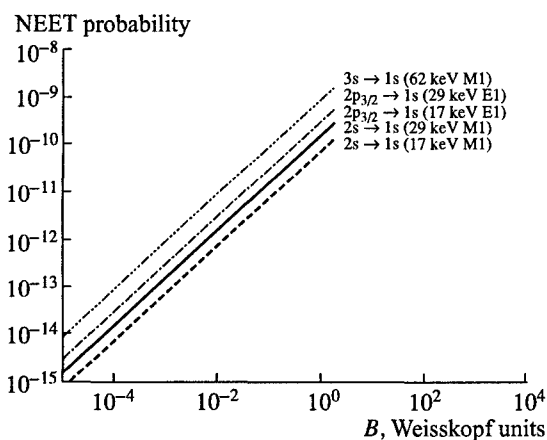
where  $\lambda_{E \rightarrow F}$  is the total (IC +  $\gamma$ ) decay rate from level  $E$  to level  $F$  and  $\lambda_{E \rightarrow I}$  is the total decay rate from level  $E$  to level  $I$ . Equation (6) assumes that the nuclear level  $E$  possesses only two decay branches.

### 3. RESULTS

According to the nuclear-level scheme in [8], only two levels lying within 80 keV of the m2 isomer (2446.1 keV above the ground level) have been fully characterized. One of these is the level lying at 2433.3 keV, which corresponds to the 13- level of the  $K = 8_1$  band. This level is populated in the 12.7-keV E3 deexcitation of the m2 isomer, its dominant decay branch. The other level lies at 2485 keV above the ground state and corresponds to the 12- level of the  $K = 8_2$  band. Such a level could be excited from the m2 level by a 39-keV M4 transition. There is some tentative data for levels lying 17, 28.6, and 62 keV above the m2 level, but none of these has a spin/parity assignment. Nevertheless, we can derive upper limits on the NEET probabilities and NEET cross sections for excitation of these levels by considering the lowest multipolarity transitions possible, namely, M1, E1, and E2 transitions. In the following, we have calculated the transition-matrix elements in Eq. (2) using Dirac-Fock electron wave functions [9]. Electronic transitions appropriate to the multipolarities considered here are  $n'\kappa' \rightarrow n\kappa$  with  $n = 1, 2$ ,  $n' = 2-5$ , and  $|\kappa| \leq 3$ , with  $\kappa$  being the relativistic quantum number. Table 1 contains a summary of the energies, widths, and photoexcitation cross sections for the orbitals of hafnium with  $n \leq 3$  based on data in [10-12].

There have been no claims of triggering near the K edge [1], but only 1s vacancies can support NEET for the known nuclear levels discussed above, due to energy conservation. In Fig. 1, we show the NEET probabilities as a function of  $B$  for several transitions filling 1s holes. The results for  $B = 1$  Weisskopf unit (W.u.) can be considered as effective upper limits for  $L = 1$  transitions, which are likely to be significantly hindered. NEET transitions with  $L > 1$  have significantly smaller probabilities than those with  $L = 1$ , even for intraband E2 transitions, for which  $B$  can attain values near 100 W.u.

For both E1 and M1 transitions, the total NEET probability for filling a 1s hole for  $B = 1$  W.u. is of the order of  $10^{-9}$  or less. The  $3s \rightarrow 1s$  transition probab-



**Fig. 1.** Probabilities for  $L = 1$  NEET transitions to nuclear levels at 17, 29, and 62 keV filling a K-shell hole. The curves for the  $2p_{3/2} \rightarrow 1s$  (17 keV E1) and  $2s \rightarrow 1s$  (29 keV M1) coincide.

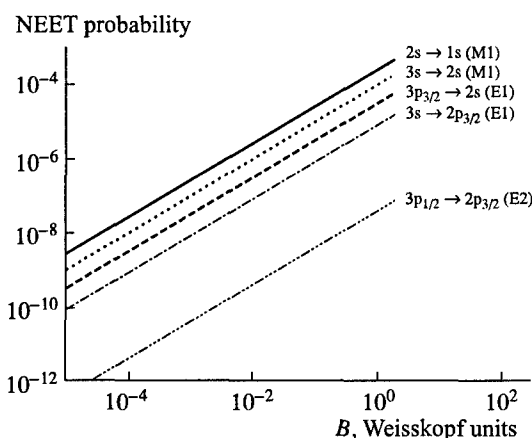


Fig. 2. Probabilities for NEET transitions filling K- and L-shell holes assuming perfect energy matching between the nuclear and electronic transitions.

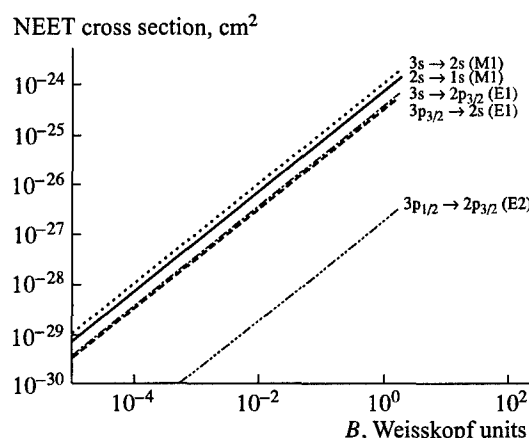


Fig. 3. Cross sections for NEET transitions filling K- and L-shell holes assuming perfect energy matching between the nuclear and electronic transitions.

ity, for the level lying 62 keV above the m2 isomer, is largest because energy matching is the closest in this case, with a detuning  $\delta = \omega_e - \omega_N$  value less than 1 keV. The other transitions are far from resonance and have energy mismatches of the order of tens of keV. Excitation of the level lying 39 keV above the m2 isomer by an M4 NEET transition has a probability of less than  $10^{-25}$  even for  $B = 1$  W.u., due to the high multipole order and the large detuning. We emphasize that the NEET probability is zero for L-shell excitations of the known nuclear levels.

We now consider NEET transitions filling L-shell holes. Since there are no known nuclear levels lying within 16 keV above the m2 isomer, such NEET transitions require hypothesizing the existence of an unknown nuclear level. We can derive upper limits on the NEET probabilities by assuming that the hypothetical nuclear level exists with an energy that would give perfect resonance with the electronic transition ( $\delta = 0$ ). NEET probabilities calculated on this basis are shown in Fig. 2. Using Eq. (4) gives the cross sections shown in Fig. 3. If we assume  $B < 1$  W.u., then we obtain an upper limit on the NEET probabilities of  $P_{\text{NEET}} < 2 \times 10^{-4}$  for transitions filling L shells. The corresponding result for transitions filling the K shell is  $P_{\text{NEET}} < 2 \times 10^{-3}$ , in agreement with previous results on the  $Z = 70$  atom [13]. These correspond to excitation cross sections of the order of  $10^{-23}$  cm<sup>2</sup>, using Eq. (4) and values from Table 1. Such cross sections are relatively large, but it must be remembered that these are extreme upper limits, since the probability of a nuclear level lying within a few keV above the isomeric level is remote. Furthermore, as discussed below, these cross sections are purely for nuclear excitation and do not take into account the process of nuclear deexcitation to lower lying nuclear levels. We can compare these cross sections with the cross sections for direct nuclear photoex-

citation. The integrated cross sections for nuclear photoexcitation for transitions at 10 keV with  $B = 1$  W.u. are  $4 \times 10^{-23}$  cm<sup>2</sup> eV for E1 transitions,  $1 \times 10^{-31}$  cm<sup>2</sup> eV for E2 transitions, and  $4 \times 10^{-25}$  cm<sup>2</sup> eV for M1 transitions. The recent experiments by Collins used narrow-bandwidth radiation with a bandwidth of the order of 0.1–1 eV. Such bandwidths are much larger than the widths for nuclear photoexcitation but much smaller than the energy scale ( $\approx$ keV) of the gross features in the above-threshold electronic photoexcitation cross section. Thus, to obtain estimates of integrated cross sections for nuclear photoexcitation by NEET, we can simply multiply the above-mentioned NEET cross sections by the beam width. For a 1-eV wide beam, the integrated cross section for NEET is  $10^{-23}$  cm<sup>2</sup> eV for the E1  $3d_{3/2} \rightarrow 2p_{3/2}$  transition,  $10^{-24}$  cm<sup>2</sup> eV for the M1  $3s \rightarrow 2s$  transition, and  $10^{-27}$  cm<sup>2</sup> eV for the E2  $3p_{3/2} \rightarrow 2p_{1/2}$  transition. Hence, for equal  $B$  values, the cross section for direct E1 photonuclear excitation is one order of magnitude larger than that for the E1 NEET transition at  $\delta = 0$ , while for the M1 and E2 transitions, the cross sections for direct photonuclear excitation are, respectively, one and four orders of magnitude smaller than those for the NEET transitions at  $\delta = 0$ .

A recent theoretical calculation on NEET excitation in  $^{178\text{m}2}\text{Hf}$  by the filling of L-shell holes was reported by Tkalya [14], who also assumed perfect energy matching between the electronic and nuclear transitions. The NEET matrix elements and probabilities obtained here are in good agreement with those reported by Tkalya: the values agree within 20% except in the case of the  $3s \rightarrow 2p_{3/2}$  transition, where the squared matrix element in the present calculation is  $1.6 \times 10^{-4}$  eV<sup>2</sup>, which is somewhat smaller than the value of  $2.7 \times 10^{-3}$  eV<sup>2</sup> reported in [14].

**Table 2.** Summary of branching ratios for the dominant nuclear excitation and deexcitation pathways

Excited level $E$	Final level $F$	$(\pi L)_{IE}$	$(\pi L)_{EF}$	$R$ ( $f=100$ )	$R$ ( $f=66$ )
15-	13-	$E1$	$E2$	$6 \times 10^{-15}$	$4 \times 10^{-14}$
14+	13-	$E2$	$E1$	$4 \times 10^{-5}$	$3 \times 10^{-4}$
15+	13-	$M1$	$M2$	$9 \times 10^{-16}$	$7 \times 10^{-15}$

In order to calculate the branching ratio for deexcitation of level  $E$  to a nuclear level other than the  $m2$  isomer, some assumptions have to be made about the type of nuclear state  $E$  produced in the NEET process. The most favorable cases for an excitation from the  $J\pi = 16+$ ,  $K = 16$   $m2$  isomer to any higher lying state would be  $E1$ ,  $M1$ , or  $E2$ . We will consider the likelihood of various angular-momenta assignments for the hypothetical level based on these multipolarities. In the case of an  $E2$  excitation due to NEET, it would be possible to reach a level having  $J = 14, 15, 16, 17$ , or  $18$ . This level is required by the assumption of perfect matching with L-shell transitions to be about 11 keV or less above the isomer and, therefore, lies no higher than about 2.457 keV. Note that triggered energy release has been claimed near both the L1 and L3 edges, namely, at 11.27 and 9.56 keV, respectively [2, 3]. Members of the  $K = 0$  ground-state band attain angular momenta through collective rotation and reach only  $J = 14$  by an excitation of 2.777 keV. The highest angular momentum achieved by rotational band members built on other intrinsic states by 2.457 keV arises in the  $J\pi = 13-$ ,  $K = 8$  level in the  $m1$ -isomer band, and this is below the  $m2$  isomer. It is clear that any hypothetical level with  $J > 13$  postulated at or below 2.457 keV would be vast, with its large angular momentum coming from a single-particle structure rather than collective rotation. It would, therefore, be an isomer as well, though none has been detected thus far.

Assuming the existence of a previously undiscovered level of high intrinsic angular momentum near 2.457 keV that can be excited from the  $m2$  isomer, the question remains as to how its decay could branch to other levels. Again, a comparison of the minimum angular momentum of the hypothetical state  $E$ ,  $J\pi = 14+$ , shows that only the  $J\pi = 13-$ ,  $K = 8$  level is available for a reasonable decay path by modest multipolarity. Reaching this proposed level  $E$  would require an  $E2$

transition, since the nuclear excitation and decay of  $E$  to the 13- level would be  $E1$ .  $M1$  or  $E2$  transitions from  $m2$  to  $E$  could produce  $J\pi = 15+$  or  $15-$ . Again, the paucity of lower lying levels of significant angular momentum suggests that an energy-releasing branch is only likely to the same 13- state. Supposing angular momenta  $J > 15$  for level  $E$  would only increase the difficulty involved in finding a suitable level to which it could decay in order to release the nuclear excitation. We reject the temptation to introduce yet another hypothetical state as a destination for such a decay branch. Thus, the most likely among improbable designations for the trigger level  $E$  would be  $J\pi = 14+$ ,  $15+$ , or  $15-$ . On this basis, characteristics for the deexcitation of  $E$  are shown in Table 2. Branching ratios have been estimated by the same procedure as in [14], using the relation  $\lambda_{EF} = \lambda_{EF}^W H$  with  $\lambda_{EF}^W$  as the Weisskopf  $B$  value, and the hindrance  $H_{EF}$  was derived from

$$H_{EF} = f^{(|K_E - K_F| - L_{EF})}, \quad (7)$$

where  $f$  is the reduced hindrance and  $L_{EF}$  is the multipolarity of the  $E \rightarrow F$  transition. In [14], a value of  $f = 100$  was assumed, which yielded branching ratios of approximately  $10^{-14}$  for  $M1$  and  $E1$  transitions and  $10^{-5}$  for  $E2$  transitions. For  $^{178}\text{Hf}$ , the  $B$  value of the dominant decay branch by the 12.7-keV  $E3$  transition of the  $m2$  isomer is reproduced with a value of  $f = 66$  [15]. This gives the values for  $R$  shown in Table 3. Using the above results together with Eq. (5), the results for the total deexcitation cross sections are shown in Table 3. The upper limit on the total deexcitation cross section for NEET transitions filling L-shell holes then becomes  $\sigma_{I \rightarrow E \rightarrow F}(L) < 10^{-31} \text{ cm}^2$ . This result is in agreement with the results of [14], where an upper limit of  $\sigma_{I \rightarrow E \rightarrow F}(L) < 10^{-32} \text{ cm}^2$  was deduced.

#### 4. CONCLUSIONS

NEET transitions filling the L shell of hafnium require the existence of an unknown nuclear level lying within 11 keV of the isomeric level. Upper limits on the NEET probability can be derived assuming a  $B$  value of 1 Weisskopf unit (W.u.) for the nuclear transition and perfect energy matching between the nuclear and electronic transitions. The extreme upper limit on the triggering cross section is  $\sigma < 10^{-31} \text{ cm}^2$ , supporting the recent results presented in [14]. This is significantly

**Table 3.** Summary of NEET probabilities and cross sections for the dominant transitions

Transition	$\pi L$	$P_{NEET}$	$P_{NEET}(\text{cm}^2)$	$R$	$\sigma_{I \rightarrow E \rightarrow F}(\text{cm}^2)$
$3s \rightarrow 2s$	$M1$	$1.1 \times 10^{-4}$	$1 \times 10^{-24}$	$10^{-14}$	$10^{-38}$
$3p_{3/2} \rightarrow 2s$	$E1$	$2.8 \times 10^{-5}$	$2 \times 10^{-25}$	$10^{-14}$	$10^{-39}$
$3s \rightarrow 2p_{3/2}$	$E1$	$8.0 \times 10^{-6}$	$4 \times 10^{-25}$	$10^{-14}$	$10^{-39}$
$3p_{3/2} \rightarrow 2p_{3/2}$	$E2$	$4.0 \times 10^{-8}$	$3 \times 10^{-27}$	$10^{-4}$	$10^{-31}$

**Table 4.** Subshell internal conversion coefficients,  $\alpha_{nk}$ , for the 12.7 keV E3 decay of  $^{178m2}\text{Hf}$ 

Subshell $n\kappa$	$\alpha_{nk}$
$2p_{1/2}$	$4.0 \times 10^6$
$2p_{3/2}$	$5.7 \times 10^6$
$3s$	$9.7 \times 10^3$
$3p_{1/2}$	$1.5 \times 10^6$
$3p_{3/2}$	$1.9 \times 10^6$
$3d_{3/2}$	$2.9 \times 10^5$
$3d_{5/2}$	$4.6 \times 10^5$
$4s$	$2.8 \times 10^3$
$4p_{1/2}$	$3.8 \times 10^5$
$4p_{3/2}$	$4.8 \times 10^5$
$4d_{3/2}$	$7.0 \times 10^4$
$4d_{5/2}$	$1.1 \times 10^5$
$5s$	$5.0 \times 10^2$
$5p_{1/2}$	$5.5 \times 10^4$
$5p_{3/2}$	$6.6 \times 10^5$
$5d_{3/2}$	$3.9 \times 10^3$
Total	$1.5 \times 10^7$

less than the cross section deduced from the experimental data in [2, 3]. Hence, it would seem that the reported  $\gamma$ -ray enhancements are far too large to be explained by NEET.

In [2, 3], the electron bridge was also mentioned as a potential mechanism for photon-induced decay of the isomer. The electron bridge is a third-order or higher contribution to the  $\gamma$ -decay rate of the nucleus and occurs when an initially excited nuclear state decays by excitation of an electron to a virtual state that deexcites to a lower lying final electronic state. Experimental data on the electron bridge is scarce. In [16], the 80.7-keV M4 decay of  $^{193m}\text{Ir}$  was examined experimentally, and it was suggested that the electron bridge constitutes approximately 20% of the  $\gamma$ -decay rate. Since the internal conversion coefficient in this case is approximately  $1.5 \times 10^5$ , this would correspond to an electron-bridge contribution to the total rate on the order of  $10^{-5}$ – $10^{-6}$  of the total rate. The energy (12.7 keV) and multipolarity (E3) for  $^{178m2}\text{Hf}$  decay are not the same as those for  $^{193m}\text{Ir}$  decay. Hence, the electron-bridge contributions could be different also. However, both decays share the characteristic that the dominant internal conversion occurs in the L shell (see Table 4). Additionally, the radial operators of the matrix elements for internal con-

version by E3 and M4 transitions are such that the region of space that gives the dominant contribution to the matrix elements is approximately the same in both cases. Hence, the electron-bridge contribution in  $^{178}\text{Hf}$  might not be too far from that in  $^{193}\text{Ir}$ . We are currently evaluating the electron-bridge contribution to  $^{178m2}\text{Hf}$  with an eye to determining the extent to which this contribution could be changed by the presence of electron holes due to photon irradiation with low-energy X rays. At the present time, the most plausible explanation of the results in [2, 3] would appear to be an anomaly in the subtraction of the background  $\gamma$ -decay spectrum from the spectrum collected during the synchrotron pulse.

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